

PATENT COOPERATION TREATY

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INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

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Applicant's or agent's file reference 11321P068WO	FOR FURTHER ACTION	
International application No. PCT/US2004/019769	International filing date (day/month/year) 21.06.2004	Priority date (day/month/year) 20.06.2003
International Patent Classification (IPC) or national classification and IPC C08K7/24, C08K3/04, C08K7/06, C01B31/02, C08F292/00		
Applicant WILLIAM MARSH RICE UNIVERSITY et al.		

1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 7 sheets, including this cover sheet.
3. This report is also accompanied by ANNEXES, comprising:
 - a. ☒ sent to the applicant and to the International Bureau a total of 3 sheets, as follows:
 - ☐ sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).
 - ☒ sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.
 - b. ☐ (sent to the International Bureau only) a total of (indicate type and number of electronic carrier(s)) , containing a sequence listing and/or tables related thereto, in computer readable form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).

4. This report contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☒ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the international application
- ☒ Box No. VIII Certain observations on the international application

Date of submission of the demand 17.01.2005	Date of completion of this report 18.11.2005
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized Officer Meiners, C Telephone No. +49 89 2399-6056



INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/US2004/019769

Box No. I Basis of the report

1. With regard to the **language**, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ This report is based on translations from the original language into the following language, which is the language of a translation furnished for the purposes of:

- ☐ international search (under Rules 12.3 and 23.1(b))
- ☐ publication of the international application (under Rule 12.4)
- ☐ international preliminary examination (under Rules 55.2 and/or 55.3)

2. With regard to the **elements*** of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report)*:

Description, Pages

1-12 as originally filed

Claims, Numbers

17-34 as originally filed
1-16 filed with telefax on 24.06.2005

Drawings, Sheets

1/5-5/5 as originally filed

☐ a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing

3. ☒ The amendments have resulted in the cancellation of:

- ☐ the description, pages
- ☒ the claims, Nos. 13
- ☐ the drawings, sheets/figs
- ☐ the sequence listing (*specify*):
- ☐ any table(s) related to sequence listing (*specify*):

4. ☒ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).

- ☐ the description, pages
- ☒ the claims, Nos. 1,10
- ☐ the drawings, sheets/figs
- ☐ the sequence listing (*specify*):
- ☐ any table(s) related to sequence listing (*specify*):

* If item 4 applies, some or all of these sheets may be marked "superseded."

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/US2004/019769

Box No. II Priority

1. ☐ This report has been established as if no priority had been claimed due to the failure to furnish within the prescribed time limit the requested:
 - ☐ copy of the earlier application whose priority has been claimed (Rule 66.7(a)).
 - ☐ translation of the earlier application whose priority has been claimed (Rule 66.7(b)).
2. ☒ This report has been established as if no priority had been claimed due to the fact that the priority claim has been found invalid (Rule 64.1). Thus for the purposes of this report, the international filing date indicated above is considered to be the relevant date.
3. Additional observations, if necessary:

Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	2-9,12,15,19,23,27,28,32,33,34
	No: Claims	1,10,11,13,14,16-18,20-22,24-26,29-31
Inventive step (IS)	Yes: Claims	-
	No: Claims	1-34
Industrial applicability (IA)	Yes: Claims	1-34
	No: Claims	-

2. Citations and explanations (Rule 70.7):

see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Re Item II.

The claimed priority (US60480348, filed 20.06.2003) has been found to be invalid for the subject-matter of original claims 1-34, since the priority document only relates to arylbromide-functionalized carbon nanotubes (CNTs) as starting compounds, and only tetrahydrofuran is used in the claimed priority document as solvent b) according to original claim 1 of the present application.

Re Item V.

Reference is made to the following documents:

- D1 : WO 02/060812 A (WILLIAM MARSH RICE UNIVERSITY; TOUR, JAMES, M; BAHR, JEFFREY, L; YANG,) 8 August 2002 (2002-08-08)
D2 : BRATCHER, MATTHEW; GERSTEN, BONNIE; JI, HELEN; MAYS, JIMMY: "Study in the Dispersion of Carbon Nanotubes" MATERIALS RESEARCH SOCIETY SYMPOSIUM PROCEEDINGS, vol. 706, 2002, pages Z9.29.1 - Z9.29.6, XP001205807
D3: US-A-3287298 (DAL MON RESEARCH) 22 November 1966 (1966-11-22)
D4: WO-A-2004/046031 (RENSSELAER POLYTECHNIC INSTITUTE) 3 June 2004 (2004-06-03)

1. Application documents

- 1.1 The amendments filed with the telefax dated 24.06.2005 introduce subject-matter which extends beyond the content of the application as filed, contrary to Article 34(2)(b) PCT. The amendments concerned are the following: In claim 1 as originally filed, it is only stated that the sidewalls of the CNTs are functionalized with aryl halides. However, in amended claim 1, it is referred to aryl halide functionalized CNTs, in general. Hence, the subject-matter of amended claim 1 can be regarded as unallowable generalization of the subject-matter of original claim 1.
- 1.2 Thus, the present international preliminary examination report is based on the subject-matter of claims 1-34 **as originally filed**.

2. Novelty (Art. 33(2) PCT)

- 2.1 Document D2 discloses two routes to carbon nanotubes (CNTs) onto which polymer chains are grafted: i) Polymer chains (e.g. polyethylenimines) can either be grafted onto reactive sites on the CNTs, or ii) the polymerization is initiated on the CNT surfaces (usually called 'grafting from' in the field). The second approach involves the preparation of initiation sites for anionic polymerization, using e.g. sec-butyllithium as base for the preparation of the CNT starting anion (see D2, page Z9.29.4, third paragraph, and figure 1). THF, functionalized CNTs, and sec-butyllithium are mixed in the reactor, and after formation of the initiator species methyl methacrylate is added. Finally, the anionic polymerization is terminated by addition of methanol. It is considered that in D2 also the sidewalls of the CNTs, bear carboxylic groups, and that the polymerizations of D2 can be initiated from these defect sites.

Thus, the subject-matter of independent claims 1 and 10 of the present application is considered to lack novelty in view of D2 (Art. 33(2) PCT).

D4 is regarded as prior art under the PCT, since the claimed priority of the present application has been found to be invalid for the subject-matter of original claims 1-34. However, D4 does not disclose the processes of present claims 1-34, since the dispersion of the functionalized CNTs prior to the addition of a base is not explicitly disclosed in D4.

3. Inventive step (Art. 33(3) PCT)

- 3.1 The document D2 is regarded as being the closest prior art to the subject-matter of claims 2 and 3. D2 discloses the anionic graft polymerization of methyl methacrylate onto CNTs. The initiator is a lithiated CNT (see figure 1 of D2).
- 3.2 Concerning claim 2, it is considered that the difference between the subject-matter of claim 2 and the subject-matter of D2 resides in the absence of aryl halide

substituents attached to the CNTs of D2. In the absence of an unexpected technical effect, the objective technical problem underlying the subject-matter of claim 2 was to provide alternative CNTs as graft base over D2, which also comprise aryl halide groups. However, aryl halide-substituted CNTs are known from D1 (see e.g. figures 1, 12, and 15 of D1). Hence, the subject-matter of claim 2 is obvious to a skilled person in view of the combined teaching of documents D1 and D2. Thus, the subject-matter of claim 2 does not meet the requirements of Art. 33(3) PCT.

- 3.3 The subject-matter of independent claim 3 differs from this known D2 in that an alternative reactive site (a metallated aryl group) is used in the method of claim 3 of the present application.
- 3.4 However, it is known in the field of anionic polymerization that metallated aryl compounds can be used as initiators for the polymerization of ethylenically unsaturated monomers and that these compounds can be obtained from aryl halogenides (see e.g. D3, column 4 lines 6 - 63).
- 3.5 Also known are CNTs having phenyl bromide or phenyl chloride moieties attached to the CNT surface (see D1: figures 1, 12, and 15; examples).
- 3.6 Thus, it is considered that the subject-matter of independent claim 3 of the present application is obvious to a person skilled in the art in view of D2 taken in combination with D1 and D3.
- 3.7 It is considered that dependent claims 4 - 9 and 11 - 34 do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of novelty and/or inventive step (Article 33(2) and (3) PCT).
- 4. Industrial applicability (Art 33(4) PCT)**
- 4.1 The subject-matter of claims 1 - 34 of the present application is industrially applicable.

Re item VIII.

5. Clarity of the claims (Art. 6 PCT)

- 5.1 Claim 1: It appears that the use of CNTs which comprise either aryl halides as functionalized species **or** alternatively species comprising a nucleation site operable for anionic or ring opening polymerization in step b) of claim 1 is intended to be claimed. Otherwise, the deprotonating agent iii) mentioned in step c) of claim 1 would have no reaction partner in the form of a deprotonable species. Thus, for the assessment of novelty, the claim has been construed as if the presence of aryl halide substituents on the CNTs were also optional in step b) of present claim 1.

Furthermore, the use of the signs (i) and (ii) for different technical features is misleading.

- 5.2 Claims 29 and 30: It is not clear what is meant by 'average chain length'.